# Frequency Metrology on single trapped ions in the weak binding limit: The $3s_{1/2}$–$3p_{3/2}$ transition in $^{24}$Mg$^+$

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(Dated: January 16, 2009)

We demonstrate a method for precision spectroscopy on trapped ions in the limit of unresolved motional sidebands. By sympathetic cooling of a chain of crystallized ions we suppress adverse temperature variations induced by the spectroscopy laser that usually lead to a distorted line profile and obtain a Voigt profile with negligible distortions. We applied the method to measure the absolute frequency of the astrophysically relevant D2 transition in single $^{24}$Mg$^+$ ions and find 1 072 082 934.33(16) MHz, a nearly 400fold improvement over previous results. Further, we find the excited state lifetime to be 3.84(10) ns.

Virtually all absolute frequency measurements on single trapped ions reported so far have been performed on narrow transitions in tightly confined ions, so that the oscillation frequency $\omega_s$ of the ion around the trap center exceeded the transition linewidth $\omega_s \gg \Gamma$. In this regime, called strong-binding limit, the absorption spectrum consists of a "carrier" and a number of motional sidebands separated by $\omega_s$. Spectroscopy of the carrier eliminates first-order Doppler- and recoil shifts [1], an important prerequisite for the tremendous accuracies achieved. However, a variety of interesting transitions exist that can hardly be studied in this regime due to their large linewidths. In astrophysics strong dipole transitions observed in quasar absorption spectra are studied to constrain possible variations of fundamental constants [2], which creates a demand for accurate laboratory reference data [3]. In nuclear physics the structure of halo nuclei is being studied via isotope shift measurements on dipole transitions [4, 5], with stringent requirements on the accuracy that so far could not be met. High precision spectroscopy outside the strong-binding limit is challenging since the spectroscopy laser induces detuning-dependent heating and cooling which distorts the line profile [6]. In fact, all previous measurements in this regime were limited by the conventional spectroscopy techniques used to overcome the heating effects (e.g. [5, 7]). In this letter we present both theory and an experimental demonstration of a new spectroscopy method that essentially removes the limitations due to the back action of the interrogating laser and allows to observe a well understood line shape with high signal-to-noise ratio. The experimental demonstration yields a resonance statistically indistinguishable from a Voigt profile and thus allows to determine the line center and -widths with unprecedented accuracy.

The basic idea is to prepare a crystallized chain of ions stored in a linear radio frequency (RF) trap which is continuously laser-cooled on one side only (see Fig. 2). Ions at the other end of the chain are sympathetically cooled but do not scatter photons from the cooling laser. A spectroscopy beam less intense than the cooling beam is then directed collinearly at the ion chain and an imaging photo detector records only photons from the sympathetically cooled ions. Temperature variations that lead to line shape distortions are strongly suppressed, and adverse effects from the cooling laser (background, ac Stark shift) are eliminated, too.

Measurement schemes involving a dedicated cooling and spectroscopy beam have previously been used for spectroscopy on extended samples of ions (Mg$^+$ in a Penning trap [6], Ca$^+$ in a Paul trap [7]). Unique to our approach is that we perform spectroscopy on individual ions with spatially separated beams which, as we will show, enables order-of-magnitude higher accuracy.

We demonstrate this method by measuring the 42 MHz wide $3s_{1/2}$–$3p_{3/2}$ transition near 280 nm in single $^{24}$Mg$^+$ ions. This line is an "anchor line" for the many-multiplet method [2] used for the search for drifts of the fine-structure constant in quasar absorption spectra and has been requested for re-measurement [3]. Our new approach allows us to determine the line center to within 160 kHz, a 375fold improvement over previous results [8, 9]. Further, this is the first demonstration of an accuracy better than 1% of the linewidth in this regime, as required for the study of halo nuclei [4]. Thanks to the well understood line shape we could determine the lifetime of the $3p_{3/2}$ state in excellent agreement with previously published values [10]. The presented measurement is the first absolute frequency measurement on a single, weakly bound ion.

To quantify how well our technique can suppress detuning-dependent temperature variations, consider two crystallized ions where one ion is cooled while spectroscopy is performed on the other. The motion can be described as a linear combination of their two eigenmodes, the center-of-mass and breathing mode. Since the motional sidebands are not resolved, the cooling laser will cool both modes simultaneously. For these conditions we calculate the equilibrium temperature as follows: The
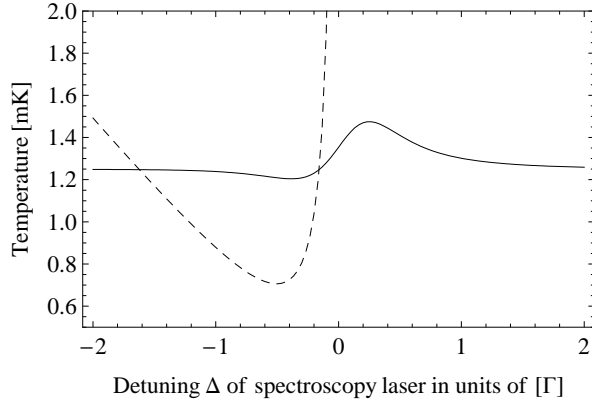
FIG. 1: Calculated temperature of the spectroscopy ion in a chain of two ions. The intensity of the cooling laser is set to a saturation parameter of $s = 0.5$ and tuned 64 MHz below resonance. The spectroscopy beam is set to $s = 0.004$. The dashed line shows for comparison the temperature profile if the cooling laser is turned off.

secular cycle-averaged cooling or heating power $\langle P_l \rangle$ due to the interaction with one of the two laser beams, enumerated by $i$, reads [11]

$$\langle P_l^i \rangle = \langle \hbar k v_0 \cos(\omega_s t)\, \Gamma g(s_i, \Delta_i - k v_0 \cos(\omega_s t)) \rangle. \quad (1)$$

Here, $\hbar$ denotes Planck's constant, $k$ the wave vector, $s_i$ is the dimensionless saturation parameter that measures the intensity in units of the saturation intensity, $\Delta_i$ the detuning, $v_0$ the velocity amplitude, $\Gamma$ the natural linewidth and $g$ finally represents the Lorentzian line shape $g(s, \Delta) = s/2[1 + s + (\frac{2\Delta}{\Gamma})^2]$. Spontaneous emission heats the motion on average by

$$\langle P_h^i \rangle = \langle (1 + \xi) \frac{\hbar^2 k^2}{2m} \Gamma g(s_i, \Delta_i - k v_0 \cos(\omega_s t)) \rangle, \quad (2)$$

where $m$ is the ion's mass and $\xi = 2/5$ for non-isotropic dipole radiation. Since in both modes the ions move with the same velocity modulus at any instant of time we can solve the steady state condition $\sum_i (\langle P_l^i \rangle + \langle P_h^i \rangle) = 0$ for $v_0$ to obtain the ion's temperature $T = m\langle v^2 \rangle / 2k_b$, where $\langle v^2 \rangle = \frac{1}{2}v_0^2$. The result of such a calculation with parameters typical for our experiment is shown in Fig. 1.

This treatment of the cooling dynamics is straight forwardly generalized to strings of $N$ ions by considering the chain mode by mode. An important difference is that the cooling laser will not cool all modes equally efficient if it interacts with a limited number of ions. For example, the stretch mode of an odd-numbered string of ions is not cooled if the cooling laser is focused on the center ion only. An analysis of the normal modes shows that it is in general favorable to cool a group of ions at one end of the chain and perform spectroscopy on ions that lie symmetrically at the other end. For such a configuration, molecular dynamics simulations show that long

ion chains are very well approximated by chains of two ions as described above if the intensities $s_i$ are chosen such that the overall ratio of cooling to heating remains the same. The cooling dynamics remains unchanged for heterogenous ion chains consisting of different species.

For a weakly bound ion $\omega_s \ll \Gamma$ in the classical limit $\hbar \omega_s \ll k_b T$, we expect the line shape to be a convolution of a power-broadened Lorentzian with a Gaussian due to the Maxwellian distribution of kinetic energies [12]. Thus, for constant temperature a Voigt profile is expected [11]. However, the residual temperature variations calculated above cause the line shape to differ slightly from a Voigt profile. To estimate size and impact of the deviations we fitted Voigt profiles to synthetic data generated according to the calculations described above. For our experimental conditions we find a systematic shift of the line center of less than 300 Hz corresponding to $< 10^{-5}$ of the linewidth. The extremely small deviations not only allow to determine the line center with great accuracy but also allow to extract information related to the linewidth precisely, e.g. the temperature of the ion and the lifetime of the excited state. The departure from a Voigt profile leads to an error determining the Lorentzian linewidth of less than 0.05 %. These errors can be reduced even further by lowering the intensity ratio.

To demonstrate the method we have measured the $3s_{1/2} - 3p_{3/2}$ transition near 280 nm in $^{24}$Mg$^+$. Ions are stored in a linear RF trap driven at 15.8 MHz with secular frequencies of 1 MHz radially and 60 kHz axially. The cooling beam encloses an angle of about $15°$ with the axis of the ion chain horizontally and $4°$ vertically, limited by the optical access of our vacuum vessel. The Doppler cooling limits for the trap's principal axes are thus $T_{min} = (0.7, 3.5, 41.6)$ mK along the ion chain, horizontally and vertically, respectively. The pressure is below $5 \times 10^{-11}$ mbar. We generate the cooling and spectroscopy beam with separately adjustable frequency and intensity at 280 nm as follows: The output of a dye laser (Coherent 699/21) near 560 nm is frequency-doubled resonantly in $\beta$-barium-borate (BBO) [13] to produce about 15 mW at 280 nm. After spatially filtering the UV beam with a pinhole it is split and passed through two double-pass acousto-optic modulators (DP-AOM) that can be tuned within 100...190 MHz, allowing us to scan the UV beam over 180 MHz. The spectroscopy beam is stabilized in intensity and spatially filtered with an additional 15 μm pinhole before it is focused onto the ions. The focused waist size in the trapping region is $w_0 \cong 200$ μm. For the cooling beam we use the output of the DP-AOM directly (no intensity stabilization and spatial filtering) and focus the light on the ions tightly with a waist of $w_0 \cong 20$ μm. Both beams are polarized linearly. To determine the absolute frequency [14], we phase-lock the dye laser to a 100 MHz repetition rate erbium fiber laser frequency comb which is referenced to a GPS-disciplined
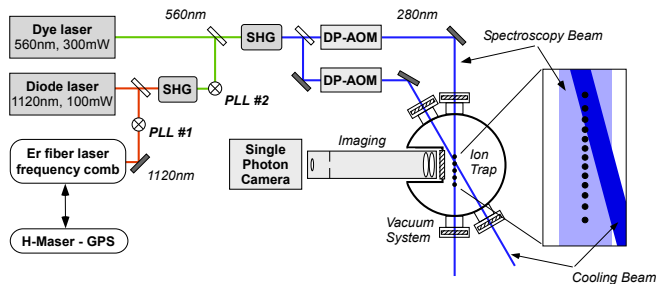
FIG. 2: Schematic of the optical setup. The dye laser is phase-locked to a fiber laser frequency comb using a diode laser as transfer oscillator (phase-locked loops PLL #1 and #2). The output of the dye laser is converted to 280 nm in a second harmonic generation (SHG) stage. Spectroscopy and cooling beams with separately adjustable frequency and stabilized intensity are obtained by splitting the UV output and passing them through two double-pass AOM (DP-AOM) setups. The inset shows the geometry of the cooling and spectroscopy beam relative to the ion chain (to scale).



FIG. 3: Typical recorded resonance. The solid line is a Voigt fit to the data, the error bars represent Poissonian noise as derived from the counts.

hydrogen maser (accuracy $10^{-14}$). A diode laser serves as transfer oscillator and bridges the gap between the comb modes at 1120 nm and the dye laser at 560 nm. We determined the mode number of the comb mode by measuring the frequency of the dye laser simultaneously with an additional frequency comb operating at a different repetition rate of 250 MHz. The optical setup is sketched in Fig. 2.

To record a line we load chains of 8-12 ions cooled at one end only by focusing the cooling laser onto 2-3 ions with an intensity of $s \cong 0.5$ and detuned 64 MHz below resonance (we did not chose $-\Gamma/2$ for technical reasons). We aligned the spectroscopy beam collinearly with the axis of the ion chain to be insensitive to possible micromotion from the radial direction. Also, this is the direction where cooling is most effective. The spectroscopy laser is set to a saturation parameter of $s \cong 7 \times 10^{-4}$. This configuration corresponds to the parameters given in Fig. 1 with six times lower spectroscopy laser intensity acting on six times more ions. We collect the fluorescence with an f/2 imaging system capable of resolving single ions (Quantar Mepsicron II, background count rate $10^{-4}$ Hz/pixel, system resolution $2\,\mu$m, total detection efficiency $6 \times 10^{-3}$). In one spectroscopy run we set the spectroscopy laser in random order to 31 different frequencies in a 180 MHz broad range centered on the transition. At each data point we collect photons for 3 s. We record the entire image, digitized in $512 \times 512$ pixels and evaluate the data by selecting circular regions-of-interest around the three outer most ions. A typical recorded line from a single ion together with a Voigt profile fitted to the data is shown in Fig. 3. In total, we recorded 264 lines from the outer three ions in 11 measurement days. The average reduced $\chi^2$ of all lines assuming Poissonian noise only is 1.1. The fit residuals show no structure other
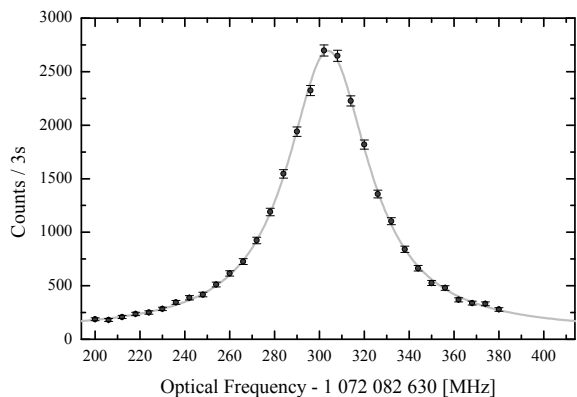
than white noise. Thus, we cannot detect a statistically significant deviation from a Voigt profile, in agreement with both our theoretical analysis and the assumption of shot-noise limited detection.

The high signal-to-noise ratio allows to separate the Lorentzian and Gaussian contributions to the width and thus to determine the lifetime of the excited state and the temperature of the ion. The average widths and statistical uncertainties of the Lorentzian and Gaussian contribution amount to 41.5(2) MHz and 11.5(3) MHz, respectively. Residual line shape distortions due to our method lead to systematic uncertainties of 14 kHz (250 kHz) for the Lorentzian (Gaussian). The low statistical and method-inherent systematic uncertainties show the potential of our approach for precision lifetime measurements, corroborated by excellent agreement with previous measurements [10]. Other systematic uncertainties not inherent to our method can be larger. With a measured upper bound on the micromotion modulation index we find from simulations that the Lorentzian and Gaussian widths are overestimated by up to 54 kHz and 170 kHz, respectively. Measurements of the polarization dependence of the linewidths indicate a systematic uncertainty due to magnetic fields of about 1 MHz. The spectrum of the dye laser available at the time of the experiment has been measured to be approximately Gaussian with a linewidth of 5 MHz and a Lorentzian contribution below 40 kHz. The width is limited by the servo loops that transfer some of the frequency comb's short term instability to the laser despite low feedback bandwidth. The resulting linewidth in the UV of 10(1) MHz gives a residual Doppler width of 6(2) MHz. This corresponds to a temperature of 1.3(8) mK, in agreement with the expected single-ion cooling limit of 1.3 mK.

To obtain an accurate line center several systematic uncertainties need to be taken into account. Static magnetic fields pointing along the spectroscopy laser shift the
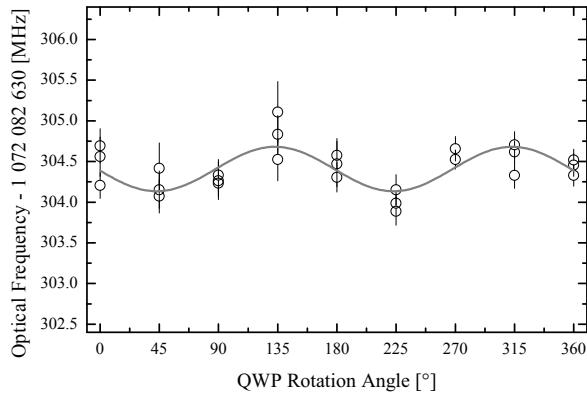
FIG. 4: Compensation of magnetic field shifts. We measured the line center versus polarization. The polarization is given in terms of the rotation angle of the quarter wave plate used; before the retarder the beam is linearly polarized. For each polarization three ions were evaluated.
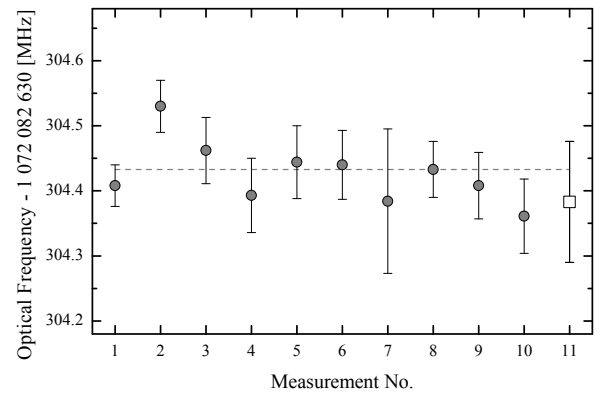


FIG. 5: Measured line centers. We determined the line center as in Fig. 4 on 11 days within four weeks. The error bars represent the statistical uncertainty only. The last data point was measured with a different laser system (fiber laser).

line center due to the linear Zeeman effect if the polarization is not perfectly linear or optical pumping takes place for other reasons. The $s_{1/2}$ ground state and the $p_{3/2}$ excited state have two and four magnetic sublevels, respectively, that are not resolved for the (laboratory) magnetic fields in our apparatus. For purely circularly polarized light the line is shifted by $14\,\mathrm{kHz}/\mu\mathrm{T}$. For this reason we pass the spectroscopy beam through a quarter wave plate retarder (QWP) and measure the line center as a function of polarization. If the polarization is expressed in terms of the rotation angle of the QWP, a sinusoidal modulation of the line center with a period of $180°$ is expected. Fig. 4 shows as an example the line centers of three ions at 9 different angles of the QWP. The amplitudes of the fitted sines correspond to a magnetic field of $19(3)\,\mu\mathrm{T}$, in agreement with a Hall probe measurement. Magnetic fields orthogonal to the spectroscopy laser should not shift the line center, which we confirmed by measuring the line center with a $200\,\mu\mathrm{T}$ field applied perpendicularly.

The DP-AOMs lead to small detuning-dependent irregular distortions of the initially Gaussian beam profile. We stabilize the total intensity but the ions sample the intensity at one point in space only, so we observe a small alignment-dependent line center shift which we remove by spatially filtering the spectroscopy beam with a pinhole. In addition, we evaluate the data by fitting a Voigt profile including a linearly varying background to account for residual intensity variations. The largest systematic uncertainty originates from the large linewidth of our spectroscopy laser and is not inherent to the method described here. By phase-locking the laser to the frequency comb we control the carrier phase. However, correlated amplitude- and phase-modulation can lead to asymmetries of the spectrum so its center-of-gravity does no longer coincide with the carrier frequency. Since the

measured line profile is a convolution of the atomic response with the laser spectrum, this can lead to systematic shifts. To estimate the size of this effect we studied the in-loop spectrum of the heterodyne beat note between the dye laser and the diode laser locked to the frequency comb. By fitting a Gaussian to the spectrum we estimated its asymmetry by determining the difference between the peak of the Gaussian and the counted heterodyne beat signal. We found an average deviation of $80(60)\,\mathrm{kHz}$ at $560\,\mathrm{nm}$, so we assume an uncertainty of $160\,\mathrm{kHz}$ in the UV. In addition we measured the transition on one day with a different laser system (a frequency-quadrupled Yb fiber laser [13]). The measurement agreed with our previous measurements (see Fig. 5), confirming our estimated uncertainty. Other systematic uncertainties we considered are significantly smaller: ac Stark shift due to residual background from the cooling laser ($30\,\mathrm{kHz}$), dc Stark shift from the trapping fields ($0.1\,\mathrm{Hz}$) [15, 16], line shape model ($270\,\mathrm{Hz}$), maser accuracy ($10\,\mathrm{Hz}$) and 2nd order Doppler shift ($-0.3\,\mathrm{Hz}$).

To test the reproducibility we repeated the measurements on eleven days within four weeks. The result is shown in Fig. 5. A weighted fit of the data gives a statistical uncertainty of $15\,\mathrm{kHz}$, with a reduced $\chi^2$ of 1. Including the correction due to the recoil shift ($-106\,\mathrm{kHz}$), the absolute frequency reads $\nu = 1\,072\,082\,934.33(16)\,\mathrm{MHz}$, in agreement with [8, 9] but 375 times more accurate.

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